

Appl. No. 10/006,231  
Amendment dated: August 8, 2003  
Reply to OA of: April 10, 2003

### **REMARKS**

Applicant has amended the specification and claims to more particularly define the invention taking into consideration the outstanding Official Action. The specification has been amended at page 1, paragraph [0004] to indicate that the white wax is paraffin white wax in accordance with the request in the Official Action. This amendment does not introduce new matter into the application but simply identifies the white wax in accordance with the chemical dictionary understanding as would be appreciated by one of ordinary skill in the art. A copy of the appropriate page from Grant & Hackh's Chemical Dictionary, page 422 is enclosed herewith. This page indicates that paraffin is synonymous with white wax. A similar amendment has been made to claim 14. In addition, claims 1-13 have been canceled from the application without prejudice or disclaimer as being directed to the non-elected invention. This amendment to the specification and claims overcomes the rejection of claims 14-24 under 35 U.S.C. 112, first paragraph. Accordingly, it is most respectfully requested that this rejection be withdrawn.

The rejection of claims 14, 17, 20-21 and 24 under 35 U.S.C. 103(a) as being unpatentable over U.S. Patent 3,560,122 to Cassar in view of JP 02/038449 to Kazuo has been carefully considered but is most respectfully traversed.

At the outset, Applicant notes that claims 15, 16, 18 and 19 are not included in the rejection and therefore these claims are free from prior art. Since the rejection under 35 U.S.C. 112 with respect to these claims has been obviated, these claims are clearly allowable and an early notification thereof is most respectfully requested.

With respect to claim 14, the Examiner notes that Cassar teaches a wax decoration item (a candle) that is made of a mixture of paraffin (which the Examiner recognizes is a white wax) and polyethylene wax with specific reference to column 2, lines 1-9. The Examiner recognizes that Cassar does not teach that the wax mixture comprises a cyanide polymer. However, the Examiner urges that JP '449 teaches a resin composition made up in part of polyethylene waxes. The resin composition is said

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to include a cyanide monomer in order to increase the antistatic properties of the resin. It is then urged that at the time of the invention, one of ordinary skill in the art would have been motivated to add cyanide to the wax mixture of Cassar with the expectation of increasing the antistatic properties of the wax emulsion. This aspect of the rejection has been carefully considered but is most respectfully traversed.

As correctly noted by the Examiner, claim 14 is directed to a wax decoration item comprising a mixture of polyethylene wax, cyanide polymer and white wax. This has been amended to specify a paraffin white wax. The Cassar reference relates to a candle wick the composition of the wick comprising paraffin wax, palygorskite particles and polyethylene. As noted at the bottom of column 2 of this patent, in order to obtain a satisfactory wick the UHMW PE and the particles must be uniformly distributed throughout the wax. This all relates to the formation of a wick which is not a decorative item of the presently claimed invention. This perhaps is more clearly seen with reference to column 4 of the patent wherein various diameter wax candles are described as being prepared from a paraffin wax. A hole was drilled in the center of each wax candle. The proper size wick in accordance with the invention therein was inserted in the hole of the body of the wax. Clearly, this teaching does not suggest the wax decoration in accordance with claim 14.

Moreover, Applicant most respectfully submits that there is no motivation to combine the references as suggested in the Official Action from the references themselves. Applicant's teaching may not be used to combine the references and arrive at the claimed invention.

JP '449 does not overcome the deficiencies of the secondary reference. JP '449 relates to a resin composition excellent in antistatic properties and dry heat discoloration resistance by mixing a aromatic polycarbonate resin with a rubber containing thermoplastic resin, a polyethylene wax and specified compounds.

It is further stated that the title composition is formed by mixing 10-95% by weight of aromatic polycarbonate resin with 5-90wt.% rubber-containing thermoplastic resin prepared by copolymerizing at least two vinyl monomers selected from among a

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methacrylic ester, an acrylic ester, an aromatic monovinyl compound and a vinyl cyanide compound in the presence of a butadiene rubber, 100 pts.wt thermoplastic resin composition formed by copolymerizing an aromatic vinyl monomer with a vinyl cyanide monomer, 1-10 pts.wt. polyethylene wax having a metal organic sulfonate group and an acidic group and 0.01-5 pts.wt. compounds of formulas I and II (wherein Y is H or a hydroxybenzyl, and at least one Y is an alkyl-substituted or unsubstituted hydroxybenzyl). This clearly in no way leads one of ordinary skill in the art to adding such a composition to that described by Cassar. Accordingly, it is most respectfully requested that this aspect of the rejection be withdrawn.

With regard to claim 17, the Examiner urges that the wax mixture of Cassar in view of JP '449 is a colloid since it is essentially a mixture of two waxes. However, claim 17 is patentable for the above reasons and further in view of the further limitations in claim 17. The Examiner's statement that the position that the wax mixture of Cassar in view of JP '449 is colloidal since it is essentially a mixture of two waxes has been carefully considered but is most respectfully traversed. This is simply an allegation and not supported by any factual evidence in support thereof. The Official Action has not established a prima facie case of obviousness in this regard. Accordingly, it is most respectfully requested that this aspect of the rejection be withdrawn.

The Official Action states that with regard to the limitation that the melting temperature and the condensing temperature of the colloid material being lower than those of the wax decoration item, that this is a design feature. However, this does not overcome the requirement that the prior art must provide the necessary motivation to one of ordinary skill in the art to make the necessary selection and basically represents nothing more than an obvious to try standard. This is not the standard for obviousness under 35 U.S.C. 103 and therefore it is most respectfully requested that this aspect of the rejection be withdrawn.

The rejection with regard to claims 20 and 21 has been carefully considered but is most respectfully submitted for the reasons as discussed above. The wax mixture referred to in Cassar relates to the wick and is not to a decorative embodiment as

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presently claimed. The unsubstantiated allegations in the Official Action with respect to the colloidal aspect of the invention does not fully consider the limitations of the claim and is unsubstantiated by any reference to the prior art. Accordingly, it is most respectfully requested that this aspect of the rejection be withdrawn. This is also true with respect to the rejection of claim 24.

The rejection of claims 22 and 23 under 35 U.S.C. 103(a) as being unpatentable over Cassar in view of JP '449 as applied to claim 14 above and further in view of the patent to Tsaor has been carefully considered but is most respectfully traversed. Contrary to the assertion in the Official Action, Tsaor and JP '449 do not suggest the claim limitations. The addition of a fragrance does not overcome the deficiencies of these references and therefore it is most respectfully requested that this rejection be withdrawn.

In view of the above comments and further amendments to the specification and claims, favorable reconsideration and allowance of all of the claims now present in the application are most respectfully requested.

Respectfully submitted,

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# GRANT & HACKH'S CHEMICAL DICTIONARY

[*American, International, European and British Usage*]

*Containing the Words Generally Used in Chemistry,  
and Many of the Terms Used in the Related  
Sciences of Physics, Medicine, Engineering,  
Biology, Pharmacy, Astrophysics,  
Agriculture, Mineralogy, etc.*

*Based on Recent Scientific Literature*

FIFTH EDITION

*Completely Revised and Edited by*

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Milan Montreal New Delhi Panama  
Paris São Paulo Singapore  
Sydney Tokyo Toronto*

**parabanic acid**  $\text{CO}\cdot\text{NH}\cdot\text{CO}\cdot\text{NH}\cdot\text{CO} = 114.1$ . Oxalylurea,

imidazolidinetrione, oxalic acid ureid. Colorless plates, m. 243 (decomp.), soluble in water. dimethyl ~ Cholestrophane.

**parabittuminous** Describing a good caking gas-coal.

**parabola** A plane curve, each point of which is equidistant from a straight line (axis) and a central point (focus). It resembles a circle at some points, a straight line at others.

**paraboloid** The solid shape traced by a parabola when rotated about the axis containing the focus. p. condenser A spherical microscope mirror having an elongated focus.

**parabuxin**  $\text{C}_{24}\text{H}_{48}\text{ON}_2 = 380.7$ . An alkaloid in common garden box, *Buxus sempervirens* (Euphorbiaceae).

**paracasein** Casein digested with rennin.

**paracellulose** Cellulose from the parenchyma or pith of plants (obsolete).

**Paracelsus (1493-1541)** Philippus Aurelius Theophrastus Paracelsus Bombastus von Hohenheim. Swiss physician and alchemist, advocate of chemical as opposed to vegetable remedies. Cf. *iatrochemists*.

**paracetamol** EP, BP name for acetaminophen.

**parachor**  $P = M\gamma^{1/4}/(D - d) = 0.78 \times V$ , where  $V$  is the critical volume,  $\gamma$  surface tension,  $M$  molecular weight, and  $D$  and  $d$  densities of a compound in the liquid and vapor state, respectively, at the same temperature. A comparison of the  $P$  of liquids is equivalent to a comparison of their molecular volumes at temperatures of equal surface tension.  $P$  is an additive constant for saturated compounds and is used to determine chemical constitution.

**Paracon** Trademark for an oil- and heat-resistant synthetic rubber consisting of chain esters of sebacic or succinic acid and ethylene or propylene glycols.

**paraconic acid**  $\text{O}\cdot\text{CH}_2\cdot\text{CH}(\text{COOH})\cdot\text{CH}_2\cdot\text{CO} = 130.1$ .

Italmic acid  $\gamma$ -lactone, tetrahydro-5-oxo-3-furancarboxylic acid. Colorless crystals, m. 58. Cf. *citraconic acid*. dimethyl ~ Terebic acid. phenyl ~ See *phenylparaconic acid*.

**paraconine**  $\text{C}_8\text{H}_{15}\text{N} = 125.2$ . An alkaloid obtained by heating butyraldehyde with ammonia. Colorless liquid with stupefying odor, b. 170.

**paracoto** The dried bark of an unidentified tree of N. Bolivia; a substitute for *coto* bark, q.v.

**paracotoin**  $\text{C}_{12}\text{H}_8\text{O}_4 = 216.2$ . An active principle from paracoto. Yellow crystals, m. 150.

**paracyanogen**  $(\text{CN})_5 = 130.1$ . (1) An insoluble solid, sublimes if heated. (2) More correctly, the water-insoluble polymer,  $(\text{CN})_n$ , of unknown molecular weight. Brown powder converted into cyanogen when heated above 860° in absence of air. Produced by prolonged pyrolysis of cyanogen at 300°.

**paradiazine** Pyrazine\*.

**paradimethylaminobenzaldehyde**  $\text{C}_6\text{H}_4(\text{CHO})\text{NMe}_2 = 149.2$ . A reagent for indole, skatole, pyrrole.

**paradioxybenzene** Hydroquinone\*.

**paraffin** (1) See *alkanes*. (2) Hard p. (BP). White wax, d. 0.890, m. 47-65, insoluble in water, soluble in organic solvents. P. is a mixture of hydrocarbons occurring native in ozocerite, peat, and bituminous coal, and is a constituent of petroleum from which it is distilled. Used in the manufacture of ointments (NF, BP), waxed paper, matches, lubricants, oil crayons; and for waterproofing wood and cork. (3) See *kerosine*. liquid ~ BP name for mineral oil. white soft ~, yellow soft ~ BP names for petrolatum.

p. bath Molten paraffin. p. oil Petrolatum. p. scale A crude paraffin. p. wax Paraffin (2).

**paraffins** Alkanes\*. iso ~ Aliphatic, saturated hydrocarbons containing one  $-\text{CHMe}-$  group or a side

chain. normal ~ Aliphatic, saturated hydrocarbons containing only  $\text{CH}_3-$  and  $-\text{CH}_2-$  groups.

**paraffinum** Paraffin.

**paraform** 1,3,5-Trioxane.

**paraformaldehyde** 1,3,5-Trioxane. Cf. *paraldehyde*.

**parafuchsin**  $(\text{C}_6\text{H}_4\text{NH}_2)_2\text{C}(\text{OH})\text{C}_6\text{H}_4\cdot\text{NH}\cdot\text{HCl}$ .

Pararosaniline chloride. A dye. Cf. *pararosaniline*.

**paragenesis** The passage of minerals through successive stages of chemical composition during the cooling of the earth's crust.

**paraglobulin** Fibr(in)oplastin. A globulin from blood serum and lymph.

**paragonite**  $\text{Al}_3\text{NaH}_2\text{Si}_3\text{O}_{12}$ . A mica-type silica mineral, q.v.

**Paraguay tea** Maté.

**parahydrogen** See *hydrogen* (2).

**paralactic acid** (S)-Lactic acid.

**paralbumin** A protein from ovarian cysts.

**paraldehyde**  $(\text{O}\cdot\text{CH}\cdot\text{Me})_3 = 132.2$ . 2,4,6-Trimethyl-1,3,5-trioxane. A polymer of acetaldehyde. Colorless liquid of pungent odor, d. 0.992, m. 11, b. 128, soluble in water; a reagent for alkaloids and fuchsin, and a hypnotic and sedative (USP, EP, BP). Cf. *metalddehyde*, *aldol*.

**paraldol**  $\text{C}_8\text{H}_{16}\text{O}_4 = 176.2$ . The dimer of aldol, m. 96.

**parallax** The apparent displacement of an object due to a change in the position of the observer, e.g., errors in buret meniscus readings.

**parallel** (1) Having the same direction, but separated by equal distances. (2) Describing electric connections such that like poles of a number of units are connected to one another. Cf. *series*.

**parallelotermism** The relationship between isomorphous groups and their chemical compositions or physical properties.

**paralyser** Paralyzer.

**paralysol**  $\text{Me}\cdot\text{C}_6\text{H}_4\cdot\text{OK}$ . A mixture of cresol and potassium cresolate. Colorless crystals, m. 146, insoluble in water; an antiseptic.

**paralyst** Paralyzer.

**paralyzant** A substance that causes paralysis.

**paralyzer** (1) An agent that prevents a chemical reaction; a catalytic poison. (2) Paralyzant.

**param**  $\text{N}:\text{C}\cdot\text{NHC}(\text{NH})\text{NH}_2 = 84.1$ . Cyanoguanidine. A condensation product of cyanamide, formed at 150, m. 204, soluble in water.

**paramagnetic** A substance that has magnetic properties stronger than those of air (as, iron); i.e., a magnetic permeability over 1. Cf. *diamagnetic*.

**paramagnetism** The property of being attracted by a magnet.

**paramecium** A genus of unicellular animals, or protozoans.

**parameter** A quantity that can be varied, but that is defined for a specific case; as, pressure.

**paramisan sodium** Aminosalicylate sodium.

**paramorph** A crystal that has undergone paramorphism.

**paramorphine** Thebaine.

**paramorphism** (1) The physical change of a mineral from one modification to another, without a change of chemical composition. (2) A rearrangement of molecular structure.

**paramucic acid**  $\text{C}_6\text{H}_{10}\text{O}_8 = 210.1$ . An isomer of mucic acid.

**paramyelin**  $\text{C}_{35}\text{H}_{75}\text{O}_9\text{NP} = 721.0$ . White solid from brain and nerve substance.

**paranaphthalene** Anthracene (obsolete).

**paranitraniline**  $\text{NO}_2\cdot\text{C}_6\text{H}_4\cdot\text{NH}_2 = 138.1$ . Yellow crystals, m. 148, soluble in alcohol; a reagent and intermediate.

**parapectic acid**  $\text{C}_{24}\text{H}_{34}\text{O}_{23} = 690.5$ . An oxyacid produced from pectose by the ripening of fruits.

**parapectone** Syntonin.

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3,560,122

## CANDLE CONTAINING WICK OF NOVEL COMPOSITION

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Int. Cl. F23d 13/16

U.S. Cl. 431-288

3 Claims

### ABSTRACT OF THE DISCLOSURE

A candle wick having the composition defined herein, and whose cross-sectional area when compared to the cross-sectional area of the candle it is contained in has a ratio between 0.042 and 0.250, is useful in place of the costly and difficult-to-make cotton wicks used in candles. The wick composition comprises 65 to 98.8 weight percent paraffin wax, 0.1 to 10 weight percent palygorskite particles and 1.1 to 25 weight percent of polyethylene having a weight average molecular weight in the range of 500,000 to 6,000,000, the latter two being uniformly distributed in the wax.

### CROSS REFERENCE TO RELATED APPLICATION

This invention concerns a novel use of the composition described in a related application Cassar application Ser. No. 840,532, filed the same date as this application.

### BACKGROUND OF THE INVENTION

The invention relates to candle having an inexpensive candle wick of novel composition in place of a conventional cotton candle wick. More specifically the invention relates to a candle containing a centrally located wick wherein the ratio of the cross-sectional area of the wick to the cross-sectional area of the candle is between 0.042 and 0.250. The composition of said wick comprises paraffin wax, palygorskite particles, and polyethylene having a weight average molecular weight in the range of 500,000 to 6,000,000, the latter two being uniformly distributed throughout the wax. The weight percent of each material is defined herein. There heretoforementioned polyethylene is also referred to herein for convenience as "USMW PE."

Much of the effort in attempting to improve the performance of candles has been directed towards improvements in the performance of cotton wicks. In Kirk and Othmer, Encyclopedia of Chemical Technology, second edition, volume 4, under "Candles" is a detailed description of the complex art involved in preparing a satisfactory wick. Birefly selected cotton yarn of certain physical properties is interwoven or plaited into a wick. This plaited wick is boiled in a weak alkali solution, bleached with chlorine or a chlorine-containing solution with hydrochloric acid and washed with clean water. Excess water is removed. A specifically prepared chemical solution is applied to the wick. After the water is removed the salts remaining improve the burning characteristics of the wick and when the flame is extinguished prevents afterglow and smoldering. After soaking or boiling in the aqueous chemical solution, the composition of which varies greatly, depending upon the manufacturer, the wick is removed, dried and inspected. The wick is then wound on spools for use.

Consequently, an inexpensive substitute which can be used in place of the heretoforementioned difficult-to-make and expensive cotton wicks in candles is highly desirable.

### SUMMARY OF THE INVENTION

The present invention is a candle containing a wick wherein the ratio of the cross-sectional area of the wick

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to the cross-sectional area of the candle is between 0.042 and 0.250. The composition of said wick comprises 65 to 98.8 weight percent paraffin wax, 0.1 to 10 weight percent of palygorskite particles and 1.1 to 25 weight percent of UHMW PE. The particles and the UHMW PE are uniformly distributed throughout the wax. The wick of said composition can be incorporated into a candle by any number of known techniques.

### DESCRIPTION

Paraffin waxes are available with a wide range of physical properties, for example, melting points may be as low as 100° F. (AMP) or as high as 160° F. (AMP). These waxes can be used in this invention. Waxes which are commercially available and would be more generally used are those having a melting point between 127° F. (AMP) and 156° F. (AMP). Other physical properties of these latter waxes are: a penetration at 77° F. of from 9 to 33 (ASTM D-1321), SUS viscosity at 210° F. of from 37.2 to 44.8 (ASTM D-446), and oil content of from 0.2 to 3.0 (ASTM D-721). AMP refers to the American melting point which is an arbitrary figure 3° F. higher than the ASTM melting point.

Polyethylenes are available with a wide range of molecular weights. These molecular weights have a substantial effect on the properties of the polyethylene. For example a polyethylene with a molecular weight of 1500 is a waxy solid while a polyethylene with a molecular weight of 1,500,000 is a hard, extremely tough solid. As discussed herein molecular weight refers to weight-average molecular weight determined by light scattering or solution viscosity or melt viscosity.

In the synthesis of polyethylene, not all the molecules grow to the same size. The actual size variation depends upon the polymerization employed. Consequently, while the molecular weight of the UHMW PE referred to in the examples herein is stated as being 1,500,000, there are in fact individual molecules in the UHMW PE with molecular weights lower than 1,500,000 and even lower than 1,000,000. Sometimes, the individual molecules have molecular weights as low as 500,000. Also in the same 1,500,000 weight average molecular weight polyethylene there are in fact individual molecules with molecular weights greater than 2,000,000, sometimes the individual molecules have molecular weights as high as 3,000,000, or even as high as 6,000,000. Thus the polyethylene used in this invention can have a weight average molecular weight in the range of 500,000 to 6,000,000.

The palygorskite particles are available in a wide range of particle sizes. However if the particles are too large the result is an extremely nonuniform composition. Thus some relatively smaller size must be used. While particles capable of passing through a 32 mesh Tyler screen can be used it is preferred that the particles be finer, e.g., capable of passing through a 60 mesh Tyler screen. Specific examples of palygorskite are attapulgite and sepiolite. These minerals are defined in Kirk and Othmer, Encyclopedia of Chemical Technology, second edition, volume 5, "Clays."

The amount of palygorskite particles used in the formulation can vary depending on the particular type of mineral and the average particle size and the particle size distribution. In the examples used for illustrative purposes 5% by weight of attapulgite was used. Generally the amount of particles in the combination will range from 0.1 to 10% by weight.

In order to obtain a satisfactory wick the UHMW PE and particles must be uniformly distributed throughout the wax. One way of achieving this uniform distribution is to add the UHMW PE as a powder and the particles, either together or separately in any sequence, to agitated molten wax, maintained at a temperature below

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the melting point of the UHMW PE and then allow the combination to solidify while being agitated. A preferred way of achieving this uniform distribution of additives in the wax is as follows. UHMW PE powder is added rapidly to the agitated molten wax containing the particles and maintained at a temperature above the melting point of the UHMW PE. As soon as the polyethylene gels the wax, the mass is allowed to cool without agitation.

After the combination of the wax, UHMW PE and the particles is prepared it can be formed into the desired shaped wick by any one of several means. For example, a circular wick can be easily formed by extrusion of either the molten formulation or solid pieces of the formulation. For illustrative purposes the wicks used in the examples were prepared by compression molding. Other means of forming the wick include casting and other types of molding.

The wick can be incorporated into the candle by any number of techniques. Thus a candle can be molded having a hollow center, the diameter of which is just sufficient for an extruded circular wick to be inserted.

Surprisingly the size of the wick used comprising the composition defined herein depends on the size of the candle. That is to say, the cross-sectional area of the wick used depends on the cross-sectional area of the candle the wick is used in. Thus the size of the formed wick used depends on the size of the body of the candle. For example, a  $\frac{1}{8}$  inch diameter cylindrical wick prepared from the formulation defined herein by itself is easily ignited and burns in a highly satisfactory manner. However if the same  $\frac{1}{8}$  inch diameter cylindrical wick is placed in a 3 inch diameter cylindrical body of wax having a  $\frac{1}{8}$  inch diameter hole located in its center a different result occurs. The wick in this candle will ignite but almost immediately thereafter it will go out. Yet a satisfactory candle is obtained by taking a  $\frac{3}{16}$  inch circular wick made from the formulation defined herein and placing it centrally within a 1 inch diameter body of wax. Thus in the former case, when the ratio of cross-sectional area of wick to the cross-sectional area of candle was 0.0418, the candle would not stay lit. In the latter case, when the ratio of cross-sectional areas was 0.1875, the candle worked satisfactorily. Thus a minimum ratio of cross-sectional area of the wick used in a candle to the cross-sectional area of the body of the candle must be obtained in order to use a wick prepared from the formulation defined here. However, as the cross-sectional area of the wick increases relative to the cross-sectional area of the candle the bottom of the flame tends to spread across the top of the candle. The disadvantage of this is that as the bottom of the flame spreads the shape of the flame more and more departs from the conventional shape of a candle flame. To control the tendency of the bottom of the flame to extend across the top of the candle the ratio of cross-sectional area of the wick used in the candle to the cross-sectional area of the candle must be less than 0.50. Thus it is distinctly preferable that this ratio of cross-sectional areas be between 0.042 and 0.25.

#### EXAMPLES

A typical paraffin wax, used herein for illustrative purposes, has a melting point of 127° F. (AMP), a viscosity at 210° F. of 43.6 SUS, a penetration at 77° F. of 33 (ASTM D-1321) and an oil content of 0.5% (ASTM D-721). A typical UHMW PE, used herein for illustrative purposes has a weight average molecular weight of 1,500,000; a specific gravity of 0.94 (ASTM D-792), a crystalline melt point of 275° F. as determined by differential thermal analysis and a nil melt index (ASTM D-1238). The palygorskite particles used herein for illustrative purposes was attapulgite. The particles of the latter were capable of passing through a 60 mesh Tyler screen. A typical chemical analysis of attapulgite yields about 55% SiO<sub>2</sub>, 10.2% Al<sub>2</sub>O<sub>3</sub>, 3.5% Fe<sub>2</sub>O<sub>3</sub>, 10.5%

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MgO, 0.5% K<sub>2</sub>O, and about 19.8% H<sub>2</sub>O removed at an elevated temperature; 99.5% total.

The compounding of the paraffin wax, UHMW PE, and clay particles was performed in the following manner. The paraffin wax was melted in a suitable container. The temperature of the molten wax was maintained at a few degrees above the melting point of the UHMW PE. The desired amount of attapulgite was added to the molten wax. While the molten wax and attapulgite mixture was being agitated the desired amount of UHMW PE powder was rapidly added. Soon thereafter the polyethylene gels the wax. After this gelation, the combination, without agitation, was allowed to solidify.

The heretofore prepared composition was compression molded into two long circular wicks. One wick has a diameter of 0.125 inch, and the other wick had a diameter of 0.1875 inch. Both of these wicks were easily ignited and remained ignited until completely consumed.

Various diameter wax candles were prepared from a paraffin wax having a melting point 145° F. (AMP), a viscosity at 210° F. of 40 SUS, a penetration at 77° F. of 9 (ASTM D-1321).

A hole was drilled in the center of each wax candle. The proper size wick was inserted in the hole of the body of wax. The test results on these various combinations of wick sizes and wax candle sizes are shown in the following table.

TABLE

Diameter of wick, inch	0.125	None	0.125	0.125	0.1875
Diameter of candle, inches		1.0	1.0	3.0	1.0
Ratio of diameter of wick to diameter of candle			0.125	0.0417	0.1875
Once ignited does it stay ignited	Yes	( <sup>1</sup> )	Yes	No	Yes

<sup>1</sup> Cannot ignite.

Thus a 0.125 inch diameter wick of the formulation defined herein ignited easily and stayed ignited. A one inch diameter candle could not be ignited by a match. When a wick with a 0.125 inch diameter was placed in a one inch diameter candle the wick was easily ignited and remained ignited until the entire body of wax was consumed. However when the same diameter wick was placed in a much larger body of wax—a candle with a 3 inch diameter—the wick could be ignited but would not stay ignited. A wick of 0.1875 inch diameter also worked satisfactorily in a 1.0 inch diameter candle.

The invention claimed is:

1. A candle containing a centrally located wick wherein the ratio of the cross-sectional area of the wick to the cross-sectional area of the candle is between 0.042 and 0.250; the composition of said wick comprising paraffin wax having uniformly distributed therein:

(a) an amount of polyethylene constituting between 1.1 and 25 weight percent of the composition and sufficient to facilitate ignitability, said polyethylene having a weight average molecular weight in the range of 500,000 to 6,000,000; and

(b) an amount of palygorskite particles constituting between 0.1 to 10 weight percent of the composition and sufficient to insure uniform distribution of the polyethylene in the wax, said particles being capable of passing through a 32 mesh Tyler screen.

2. A composition according to claim 1 wherein the wax has a melting point between 127° F. and 156° F. (AMP).

3. A composition according to claim 1 wherein the palygorskite is attapulgite.

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CARROLL B. DORITY, JR., Primary Examiner

U.S. Cl. X.R.

44—7.5



1

2

3,565,819

PROCESS OF PRODUCING MICROCAPSULES  
Karl G. Gragger, Laurel, Md., assignor to Westvaco  
Corporation, New York, N.Y., a corporation of Dela-  
ware

No Drawing. Filed Feb. 13, 1969, Ser. No. 799,105  
Int. Cl. A61k 9/04; B01j 13/02; B44d 1/02  
U.S. Cl. 252—316

8 Claims

## ABSTRACT OF THE DISCLOSURE

Pressure-rupturable microcapsules having outer walls of wax which encase droplets of an oily material are produced in the absence of coacervation forces by emulsifying an oil-in-water type of emulsion into melted wax, to form a water-in-oil type of emulsion, then dispersing the wax emulsion to form micro-droplets thereof in an aqueous phase, and cooling the resulting dispersion to solidify the wax. A membrane of hydrophilic colloid material impermeable to the encapsulated oil is formed between the oil and the wax and acts as a protective colloid. The dispersion of microcapsules can be coated onto an appropriate base sheet or the microcapsules can be recovered by spray drying or by filtering and drying.

## BRIEF SUMMARY OF THE INVENTION

This invention relates to microcapsules and to methods of making the same. More specifically, this invention relates to processes for making microcapsules, in the absence of coacervation, by several emulsification steps which result in the formation of microcapsules having walls of wax which encase an oily core surrounded by a protective colloid, the latter separating the wax and oil.

It is known that microcapsules containing oily materials can be made by various procedures. In one well-known type of process, oil droplets are encased in a complex of hydrophilic colloids by coacervation forces, and the walls of the so-obtained capsules are subsequently hardened, such as by treatment with an aldehyde. In other processes, microcapsules are formed by interfacial polycondensation, centrifugal microencapsulation, or fluidized bed encapsulation, all of which require one or more of the following procedures to produce the final product: chemical reaction, use of solvents other than water, or wall-hardening chemical reactions. None of these procedures are required when microcapsules are made according to the present invention.

In the past, the use of waxes as microcapsule wall-forming materials has had the disadvantage that waxes are readily dissolved by most organic oils, and the wax capsule walls leaked when organic oils were encased therein. Because of this problem, the use of waxes as wall-forming materials has been limited in the past to encapsulating polar liquids only. Yet, waxes offer many advantages as potential capsule wall-forming materials because they are substantially inert and are relatively inexpensive. According to the present invention, waxes can be used as capsule wall material for encapsulating organic oils as well as other water-insoluble organic liquids.

It has been discovered that microcapsules can be formed, in the absence of coacervation or any other known encapsulation techniques, by a series of emulsifications. First, an oil-in-water type of emulsion is formed by dispersing an oily material in an aqueous solution of a hydrophilic colloid material. This emulsion is then added to melted wax under constant stirring to form a water-in-oil type of emulsion, referred to hereinafter as wax emulsion. At this point, a stabilizing or emulsifying agent, such as a fatty acid, is added to the wax emulsion to aid in the subsequent dispersion of the wax emulsion in an aqueous

medium. The wax emulsion is then dispersed in an aqueous medium, which may be water alone, to form droplets of wax encasing the oil-in-water emulsion. The dispersion is then cooled to solidify the wax and produce a dispersion of microcapsules which have outer walls of wax encasing oil and the hydrophilic colloid. The hydrophilic colloid acts as an impermeable membrane to the oil and prevents the oil from plasticizing the wax capsule walls, and thus prevents the leakage problem attendant with past uses of wax for capsule walls. The microcapsules can be recovered by filtering the capsules and drying them or by spray drying the dispersion of capsules, to produce a free-flowing powder, or the dispersion can be applied as a coating to an appropriate flexible substrate, such as paper or film, and then the coated substrate can be dried.

In one embodiment of the invention, the aqueous phase into which the wax emulsion is dispersed is an aqueous solution of a binder or film-former, such as starch, gelatin, gum arabic, protein such as casein, or carboxymethylcellulose. The resulting dispersion of microcapsules, comprising dissolved binder having dispersed therein microcapsules having wax walls encasing the oil-in-water emulsion, may be coated directly on the surface of a suitable base by an appropriate coating device, such as a rod, blade, or air knife coater. The coated paper is then dried to produce paper coated with a coating of microcapsules, the binder acting to hold the capsules to the base sheet. The coating of microcapsules is pressure-responsive, as the capsules will rupture upon the application of localized pressure.

Capsules made according to this invention find utility in various fields. So-called pressure-sensitive coatings for no-carbon-required copy paper can be made according to the present invention. For example, a potential color-forming organic compound, known as a color-precursor, or combinations thereof, can be dissolved in the oil phase of the initial oil-in-water emulsion. After preparing a dispersion of microcapsules which are formed by several emulsification steps, as described above, paper is coated with the dispersion containing microcapsules of wax encapsulating the oil, color-precursor, and protective colloid, and the so-coated paper is then dried. The coated side represents the under side of a top sheet of paper. The top side of a second sheet of paper is coated with a color-forming reactant, such as attapulgite or bentonite clay. When the sheets are placed together, and localized pressure is applied to the top sheet, the capsules rupture and release the encased materials, and a color-forming reaction takes place on the attapulgite or bentonite-coated surface of the second sheet of paper.

The capsules and dispersions of capsules made according to the present invention can be used in other fields. For example, adhesive-activating materials, odoriferous compositions, food flavorings, or medicines can be encased in capsules made by the processes described herein. Thus, an adhesive-activating material can be encapsulated by our processes and the capsules can be applied to a suitable substrate as a pressure-responsive coating, with the result that the adhesive-activating material is released when the capsules are ruptured.

The internal phase of the initial oil-in-water type of emulsion for purposes of this invention is preferably a water-insoluble oily, organic material. The characteristics of the internal phase are not critical except as to their property of being insoluble in the aqueous external phase. The internal phase is an "oil" and is present in the external phase as colloidal droplets, such as is true for oil-in-water emulsions. Oils that can be used are of an infinite variety and number and include such materials as mineral, vegetable, animal, fish and synthetic oils such as castor oil, petroleum oils, and chlorinated di- and triphenyls and mixtures thereof. The oil may contain other oil-soluble

materials. If the final dispersion of capsules is to be used as a coating for no-carbon-required paper, then the oil will also have dissolved therein an oil-soluble color-precursor.

The external phase of the initial oil-in-water emulsion is an aqueous solution of a hydrophilic colloidal material which is not miscible with the internal phase. A hydrophilic colloidal material, which will form as a protective colloid between the internal oily phase and the wax capsule wall, is used. Preferably, the hydrophilic colloid material is gellable and film-forming so that when the wax microcapsule walls are solidified by cooling, the encapsulated hydrophilic colloid material becomes gelled and forms an impermeable film between the encased oil and the wax walls. It has been found that gelatin or gum arabic provides an excellent membrane impermeable to organic oils. Thus, the internal oil phase is emulsified into an aqueous solution of gelatin or gum arabic to form an oil-in-water emulsion. This emulsion is then added to melted wax, as for example microcrystalline waxes or paraffin waxes, and the oil-in-water emulsion is emulsified into the melted wax to produce a water-in-oil type of emulsion. The water-in-oil emulsion is then dispersed or emulsified into an aqueous phase which may be water or an aqueous solution of a binder, to produce microdroplets of the wax emulsion in the aqueous phase. The wax walls of the droplets solidify and the hydrophilic colloid material gels as the dispersion cools, to form microcapsules as described above. The microcapsules are pressure-rupturable to release the encased oil and any materials that are dissolved in the oil.

Emulsions for purposes of the present invention may be made by any of the usual emulsification equipment, such as by mixing the internal and external phases in a blender, colloid mill, or emulsifier. Under high shear, colloidal droplets of the internal phase are dispersed in the external phase.

#### DETAILED DESCRIPTION

The invention will be described in greater detail with the aid of the following examples.

##### Example 1

An external phase for an emulsion was prepared by dissolving 5 grams of purified pigskin gelatin, having an isoelectric point of about 9.2, in 50 milliliters of water heated to approximately 140° F. The pH of the resulting gelatin solution was about 5.4.

An internal phase for the emulsion was prepared by heating 96 grams of a mixture of chlorinated bi- and triphenyls (Aroclor 1242) to a temperature of about 140° F. An emulsion was then prepared by dispersing the internal phase into the external phase in a blender under high shear conditions, to form an oil-in-water emulsion. The resulting emulsion was stirred under gentle agitation for about 3 hours at a temperature of about 160° F., a temperature higher than the gelation point of the gelatin, to age the emulsion.

Seventy-three grams of white microcrystalline wax (163/169) were heated to about 165° F. to melt the wax. Under constant agitation, and while maintaining the temperature of the wax at about 165° F., the oil-in-water emulsion, prepared above, was slowly added to the molten wax, to form an oil-in-water-in-wax emulsion. Then about 0.5% of stearic acid, based on the weight of wax, was added to the emulsion to aid in the subsequent dispersion of the oil-in-water-in-wax emulsion in an aqueous medium.

The resulting water-in-oil type of emulsion from above was slowly added, under constant agitation, to 1,000 milliliters of a 10 percent by weight solution of carboxymethylcellulose (CMC) heated to about 165° F., to form an oil-in-water type of emulsion. In other words, droplets of wax were dispersed in the CMC solution, and the wax droplets had dispersed therein the initial oil-in-water emul-

sion of Aroclor in aqueous gelation. The dispersion of wax droplets was then allowed to cool to room temperature and the wax solidified to form discrete microcapsules dispersed in the CMC solution. The resulting dispersion of microcapsules was coated directly onto paper. Upon drying, the coated paper could be impinged by the application of pressure, causing the microcapsules to rupture and release the encapsulated Aroclor at the point of pressure application.

##### Example 2

This is an example of preparing microcapsules for use in a coating for no-carbon-required copy paper.

Example 1 was repeated, except that color-precursors were dissolved in the oil phase of the initial emulsion. In this case, the internal phase for the initial emulsion comprised 96 grams Aroclor and dissolved therein were 2 grams of each of the color-precursors 3,3-bis(4-diethylaminophenyl)-6-dimethylamino phthalide and benzoyl leucomethylene blue.

A dispersion of microcapsules was made according to the procedures described in Example 1. The microcapsules comprised walls of wax which encased the gelatin protective colloid oil (Aroclor), and the dissolved color-precursors. The dispersion of microcapsules was coated onto paper and dried. A sheet of the so-coated and dried paper was placed on top of a sheet of paper coated with attapulgite clay, an electron acceptive adsorbent, with the pressure-responsive coating on the top sheet in contact with the attapulgite clay coating on the bottom sheet. When pressure was applied to the top sheet, as by writing with a pen or pencil, the microcapsules ruptured and a color-forming reaction occurred when the color-precursors were adsorbed onto the attapulgite clay surface. After storing the pressure-sensitive coated paper for a period of about one month, there was every indication that the microcapsules were substantially leak-proof.

More than one color-precursor dissolved in the encapsulated oil is not required in order to obtain a color-forming reaction. The two chosen for purposes of illustration here were used together in order to produce an instant color-forming reaction as well as a permanent one.

As stated previously, waxes other than microcrystalline waxes may be employed in the present invention. Petroleum waxes perform well in the invention. Also, an aqueous solution of a binder other than carboxymethylcellulose may be used such as aqueous dispersions of casein, starch, gelatin, gum arabic, and the like. Or the final aqueous phase into which the wax emulsion is dispersed may not contain any binder, but may be water alone in which the formed microcapsules are dispersed for subsequent recovery. Furthermore, it is obvious that changes in the proportions of ingredients and ratios among the various internal and external phases described can be made.

From the above, it can be seen that it is possible according to the present invention to encase organic liquids other than polar ones in microcapsules having outer walls of wax which are protected from the encapsulated liquid by the expedient of a protective colloid. The microcapsules are produced by physical methods of encapsulation, rather than by chemical ones, and the microcapsules so-produced find utility in various fields, one being the pressure-sensitive coating field.

As one skilled in the art will readily appreciate, various modifications may be made in the examples and descriptions as set out above without departing from the spirit of the invention or the scope of the appended claims.

I claim:

1. The method of producing pressure-rupturable microscopic oil-containing capsules each having an oily core, an outer wall of wax, and a hydrophilic colloidal material separating the oily core from the wax wall, which comprises the steps of:

(a) dispersing a water-insoluble organic liquid in an aqueous solution of a hydrophilic film-forming col-

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loidal material, to form an oil-in-water emulsion,  
 (b) adding under agitation the oil-in-water emulsion to  
 melted wax to form an oil-in-water-in-wax emulsion  
 comprising droplets of the oil-in-water emulsion of  
 step (a) above dispersed in the melted wax,  
 (c) adding to the oil-in-water-in-wax emulsion an emul-  
 sifying agent to enable emulsification of the oil-in-  
 water-in-wax emulsion into an aqueous medium,  
 (d) dispersing the oil-in-water-in-wax emulsion in an  
 aqueous medium as droplets of wax having emulsified  
 therein the oil-in-water emulsion of step (a) above,  
 (e) and solidifying the wax droplets to form micro-  
 capsules having outer walls of solidified wax encas-  
 ing the oil-in-water emulsion of step (a) above,  
 whereby hydrophilic film-forming colloidal material  
 separates the encased water-insoluble organic liquid  
 from the wax walls of the microcapsules.

2. The method of claim 1 which includes the step of  
 dissolving an oil-soluble color-precursor in the water-  
 insoluble organic liquid before the water-insoluble organic  
 liquid is dispersed in the aqueous solution of hydrophilic  
 film-forming colloidal material.

3. The method of claim 1 in which the aqueous med-  
 ium in step (d) thereof is an aqueous solution of a film-  
 forming binder material, and the microcapsules formed  
 according to claim 1 are dispersed in the aqueous film-  
 forming binder material.

4. The method of claim 1 wherein the hydrophilic film-  
 forming colloidal material is selected from the group  
 consisting of gelatin and gum arabic.

5. The method of claim 1 wherein the hydrophilic  
 film-forming colloidal material is gelatin.

6. The method of claim 1 wherein the emulsifying  
 agent added in step (c) thereof is a fatty acid.

7. The method of claim 1 wherein the wax is a mate-  
 rial selected from the group consisting of microcrystalline  
 waxes and paraffin waxes.

8. The method of producing a dispersion of pressure-  
 rupturable oil-containing microcapsules each having an  
 oily nucleus and outer wall of wax, with hydrophilic col-  
 loidal material impermeable to the oily nucleus sep-  
 arating the nucleus and the wax wall, which comprises the  
 steps of:

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(a) dispersing a water-insoluble organic liquid in an  
 aqueous solution of a gellable hydrophilic film-form-  
 ing colloid material heated above the gelation tem-  
 perature of the said colloid material, to form an oil-  
 in-water emulsion comprising droplets of the organic  
 liquid dispersed in the aqueous solution of colloid  
 material,

(b) preparing an external phase for an emulsion by  
 heating wax above its melting point to form molten  
 wax,

(c) dispersing the oil-in-water emulsion of step (a)  
 above as droplets in the molten wax to form an oil-  
 in-water-in-wax emulsion,

(d) adding to the oil-in-water-in-wax emulsion an emul-  
 sifying agent to aid subsequent dispersion of the oil-  
 in-water-in-wax emulsion in an aqueous medium,

(e) dispersing the oil-in-water-in-wax emulsion as drop-  
 lets in an aqueous solution of film-forming binder  
 material heated above the melting point of the wax,  
 said droplets being droplets of wax having emulsified  
 therein the oil-in-water emulsion of step (a) above,

(f) and solidifying the wax and gelling the hydrophilic  
 colloid material by cooling to form microcapsules  
 having outer walls of solidified wax encasing the  
 gelled hydrophilic colloid material and organic liquid,  
 the gelled hydrophilic colloid material separating the  
 organic liquid and the wax, the microcapsules so-  
 formed being dispersed in the aqueous film-forming  
 binder material.

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